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(54) Process and apparatus for coating on printing cylinders

(57) A non-impact process and apparatus for coating printing cylinders with layers of a coating liquid, es-

pecially for coating flexographic printing sleeves with infrared sensitive layers.

surface of the coating roll. Both surfaces do not touch. The gap between them is filled with the coating liquid. Direct contact between the coating roll and the printing cylinder would destroy the surface of the sensitive photopolymerizable layer on the cylinder. This is avoided by adjusting a gap just sufficient to wet the surface with the coating liquid. Therefore, due to this gap the printing cylinder remains free from scratches or mechanical damage. The width of the coating gap is about 30 - 800 µm, preferably about 40 - 160 µm. Especially, coating gaps with a width of 50 - 100 µm are suitable. In this coating gap, the fluid film of the coating liquid splits into two parts. One part of the coating liquid spreads on the printing cylinder surface and adheres there. The other part remains on the coating roll surface. By the varying the width of the coating roll, it is possible to meter the fluid film.

[0018] The uniformity and thickness of the coating can be controlled by rotation of the printing cylinder, rotation of the coating roll and linear speed of a coating table, which supports the coating roll and the container for the coating liquid. Faster roll rotation results in higher wet coating weight. The method coats very clean edges at coating start and end of the cylinder simple by moving the coating roll up and down for start and finish.

[0019] Preferably, the coating liquid is applied overlapping for complete coverage and rapid levelling. After levelling and drying, a uniform layer on the printing cylinder is provided. Especially by spirally applying of the coating liquid, it is possible to provide a uniform, overlapping coating on the printing cylinder. Only one coating pass is needed which makes the whole coating process very fast and convenient. No change in coating weight along the length of the printing cylinder are observed; uneven coatings which are a problem with ring coating are avoided. Then the coating trail levels out and dries. As the printing cylinder rotates during drying, the coating dries much faster than on a non-rotating printing cylinder as with ring coating. The most convenient drying is under ambient conditions at temperatures of 18-24 °C and relative humidities of 20 - 80 % (room climate conditions). Relative humidities of 30 - 60 % are preferred for faster drying. Also, to speed up the drying time, hot air can be applied, e.g. air temperatures of 24-40 °C can be used. If no hot air is available and drying is under ambient conditions the air can be moved or circulated or the printing cylinder can be rotated at higher speed than during the coating process. To avoid dirt and dust contamination of the coated layer during the coating or drying cycle the air should be filtered. After drying, a very uniform thin coating is obtained on the printing cylinder.

[0020] In a similar way, multiple layers can be applied by overcoating a pre-coated layer in a second pass. If needed, different coating solutions can be coated by repeating the coating and drying cycles. Different coating weights can be adjusted by changing the coating roll rotation accordingly. Multiple coatings are beneficial if additional functional layers have to be integrated underneath or on top of the digitally imageable infrared sensitive layer, e.g. a release layer underneath the digitally imageable infrared sensitive layer for improved ablation and low stain and/or higher laser sensitivity and faster laser imaging. These features are important for customers convenience and high-quality flexographic printing forms.

Coating roll

[0021] The coating roll of this invention is preferably made of hard, non-elastic materials, such as, for example, specific plastics, metals, and ceramic materials. Furthermore, the coating roll material must be insoluble in and resistant to the organic solvents used for the coating liquid, like alcohols, esters, ketones, aromatic and aliphatic hydrocarbons. Sponge like and foamed materials are not suitable, because these materials are very sensitive to penetration and swelling of organic solvents. As a consequence of swelling, these roll materials will change their geometry, especially their diameter. This makes them unusable for the precision coating process of the present invention as the gap will change depending on the solvent uptake. Coating rolls made of such materials do not run round due to change and deformation of their roll geometry.

[0022] Typically, the coating roll materials which are suitable for the present coating process have a Shore D hardness measured according to ASTM D 2240 of at least 60, preferably of at least 70. Preferred are metals, like stainless steel and thermoplastic and thermosetting non-elastomeric polymers, like polyethylene, polypropylene, polyamides, polyesters, polycarbonates, polyurethane resins, ABS resins, polyacrylates, and polymethacrylates. Typically used are polyamides, like Nylon®, and polyesters, like polyethylene terephthalate. The surface of the coating roll can be smooth or can carry a screen or a line pattern. Also, a coating roll with gravure patterns, as known in the art of gravure printing, can be used. The fluid film transported to the printing cylinder can be metered by using such patterned surfaces. Typically, the coating roll has a roll diameter of 60 - 200 mm, preferably 90 - 150 mm. Especially, coating rolls with a diameter of 100 - 120 mm are used. Typically, the coating roll has a roll width of 5 - 50 mm, preferably 10 - 40 mm. Especially, coating rolls with a width of 20 - 30 mm are used.

Printing cylinder

[0023] In general, all kinds of printing cylinders may be coated by the process of the present invention. Preferably printing cylinders from metal or plastic covered with a seamless photopolymerizable printing layer may be used. Es-

pecially, seamless elastomeric flexographic printing cylinders and sleeves can be used. Sleeves are hollow cylinders usually made of plastic materials. These sleeves are readily and repeatably mounted and dismounted from printing drums, preferably by being expandable using pressurized air. Non-elastic, transparent or non-transparent base sleeves can carry additional layers for enhancing printing quality. Typical sleeves which can be used within the meaning of the present invention are disclosed in EP 0 696. Seamless printing cylinders and seamless printing sleeves are usually made by wrapping a flat photopolymerizable printing plate around a printing drum or a sleeve, and joining the edges together to form a seamless, continuous element. Such a process is disclosed in DE 28 44 426.

[0024] The photopolymerizable layers of such flexographic printing cylinders are prepared from known photopolymerizable materials. All photopolymerizable materials of the state of the art can be used. They usually comprise at least one elastomeric binder, at least one photopolymerizable, ethylenically unsaturated monomer, and at least one photoinitiator or photoinitiator system.

[0025] Examples of elastomeric binders are polyalkadienes, alkadiene/acrylonitrile copolymers; ethylene/propylene/alkadiene copolymers; ethylene/(meth)acrylic acid/(meth)acrylate copolymers; and thermoplastic, elastomeric block copolymers of styrene, butadiene, or isoprene. Linear and radial thermoplastic, elastomeric block copolymers of styrene and butadiene or isoprene are preferred. The quantity of binder is preferably 65 % by weight, relative to the total weight of the photopolymerizable material.

[0026] Useful monomers are the conventional ethylenically unsaturated, copolymerizable, organic compounds, such as, for example, acrylates and methacrylates of monovalent or polyvalent alcohols; (meth)acrylamides; vinyl ethers and vinyl esters; etc., in particular acrylic and/or methacrylic of butanediol, hexanediol, diethylene glykol, trimethylol propane, pentaerythritol, etc.; and mixtures of such compounds. The monomer quantity is preferably at least 5 % by weight, relative to the total weight of the photopolymerizable material.

[0027] Suitable photoinitiators are individual photoinitiators or photoinitiator systems, such as, for example, benzoin derivatives, benzil acetals, diarylphosphine oxides, etc., also mixed with triphenyl phosphine, tertiary amines, etc. The quantity of photoinitiator is usually 0.001 - 10 % by weight, relative to the total weight of the photopolymerizable material.

[0028] In addition to the main components described in the foregoing, the photopolymerizable compositions may comprise conventional additives like, for example, UV absorbers, thermal stabilizers, plasticizers, and fillers.

[0029] Especially preferred are the materials disclosed in US 4,323,637; US 4,427,759; and US 4,894,315.

[0030] Additional layers may be present on top of the photopolymerizable material. Especially barrier layers like those described in EP 0 654 150 are used. Such barrier layers comprise layers which are insensitive to actinic radiation and also such layers which are photosensitive themselves. Examples for the first type of barrier layers are those materials which are conventionally used as release layers, such as, for example, polyamides, polyvinyl alcohols, copolymers of ethylene and vinyl acetate, etc. Polyamides are especially preferred. Examples for the second type of barrier layers are photosensitive layers, comprising preferably an elastomeric binder, a monomer, and a photoinitiator, or such layers which become photosensitive when contacted with the photopolymerizable layer and which comprise an elastomeric binder and optionally fillers or other additives, but no monomer. Suitable layers are those disclosed as elastomeric layers in the multilayer cover element described in US 4,427,759 and US 4,460,675.

[0031] A protective coversheet may be on the photopolymerizable layer and removed prior to application coating of the infrared sensitive material. After the infrared sensitive material is coated on the photopolymerizable layer, the printing cylinder may further include a removable coversheet to protect the outermost layer, i.e. infrared sensitive layer.

Coating Liquid

[0032] All kinds of coating materials can be used as coating liquids in the process of the present invention such as, for example, protective materials, infrared sensitive materials, materials curable by exposure to ultraviolet radiation, etc. Preferably infrared sensitive, especially infrared ablative materials, can be applied to printing cylinders or sleeves by this process. Layers resulting from such materials can be laser imaged resulting in an integrated photomask for the printing cylinder.

[0033] The preferred infrared sensitive materials are soluble or dispersible in a developer, opaque to ultraviolet or visible light, that is, has an optical density about at least 2.5, and can be imaged with an infrared laser. These materials comprise compounds having high infrared absorption in the wavelength range between 750 and 20,000 nm, such as for example, polysubstituted phthalocyanine compounds, cyanine dyes, merocyanine dyes, etc., inorganic pigments, such as, for example, carbon black, graphite, copper chromite, chromium dioxide, etc., or metals, such as, for example, aluminium, copper, etc. The quantity of infrared absorbing compound is usually 0.1 - 50 % by weight, relative to the total weight of the material. To achieve the optical density of about at least 2.5 with actinic radiation, the infrared sensitive materials contain a compound that prevents the transmission of actinic radiation, such as, for example, dyes, organic ultraviolet absorbers such as, for example, hydroxybenzophenones, hydroxyphenylbenzotriazoles, hydroxyphenyl-s-triazines, oxalanalides, etc. or pigments, in particular the aforesaid inorganic pigments like carbon black, graphite, titanium dioxide, zinc oxide, etc. The quantity of this compound is usually 1 - 70 % by weight relative to the total weight

of the material. The infrared sensitive material contains optionally a polymeric binder, such as, for example, nitrocellulose, cellulose acetate butyrate, polyvinyl butyrate, polyurethanes, polyvinyl acetates, homopolymers or copolymers of acrylates, methacrylates, and styrenes, polyamides, polyvinyl alcohols, thermoplastic elastomeric polymers like linear and radial block copolymers of styrene and butadiene or isoprene, cyclic rubbers, etc. Other auxiliary agents, such as, for example, plasticizers, levelling agents, defoaming agents, viscosity builders, substrate wetting additives, anti-blocking additives, pigment dispersants, slip additives, etc. are possible. These compounds may be solved in conventional solvents. Typically solvents like water, alcohols, esters, ketones, hydrocarbons, or mixtures thereof are used. Suitable infrared sensitive materials are those disclosed in WO 94/03838 and WO 94/03839.

[0034] The infrared sensitive materials are usually coated onto the photopolymerizable layer of the printing cylinder or sleeve by the process of the present invention. It is also possible to coat it onto an elastomeric layer or onto a release layer as described above. By the process of the present invention, infrared sensitive layers can be coated on tacky photopolymerizable surfaces with excellent uniformity and no surface scratches or other damages. The variations of optical densities (OD) in the range of OD = 2.50 - 5.00 are better than +/- 5 % from start to end of the printing cylinder. This corresponds to typical dry coating weights in the range of 5 - 50 mg/dm², preferably 20 - 40 mg / dm² with corresponding high uniformities required for a clean laser ablation and, consequently, needed for a high flexographic print quality

Detailed description of process and apparatus

[0035] A coating head of an apparatus of the present invention consists of the coating roll described above and a container for the coating liquid, both mounted on a coating table. The coating table can be moved by controller driven motors in lateral direction, left and right, parallel to the horizontal axis of the printing cylinder. Further, it can be moved by motors up and down, so that the coating roll can be moved upwards or downwards relative to the printing cylinder surface. The coating roll dips with the lower part into the coating liquid which is filled into the container.

[0036] The complete coating process consists of a measuring cycle, a positioning cycle, a coating cycle and a drying cycle. During the measuring cycle a software program determines via two high precision fibre optical sensors the relative position of the cylinder surface versus the surface of the coating roll and in addition the start point for coating on the printing cylinder. Then the software controlled motors move the coating head into the coating start position: left or right to the point of coating start and up so that between the surface of the coating roll and the outer surface of the printing cylinder a pre-determined coating gap of 30 - 800 µm is maintained during the coating cycle. The coating is applied by simultaneous rotation of printing cylinder and coating roll and horizontal movement of the coating head. All speeds are pre-adjusted and maintained at high precision during the coating cycle. As the coating result, a more or less overlapping spiral is coated on the printing cylinder. When the coating is completed the coating head moves down and the drying continues, optional at a faster printing cylinder rotation. The coated layer is coated as a spiral resulting from the combination of printing cylinder rotation and linear coating head (coating roll) movement along and parallel to the printing cylinder axis. Basically, the coating spiral can be too wide (less than 100% coverage), trail beside trail (100 % coverage) or too narrow (overlapping of neighbouring trails). The coating spiral is too wide with uncovered areas when the coating head speed is fast and/or the printing cylinder rotation is slow or too slow for a given linear coating head speed. The spiral has to be coated in a way that the coating trail will not leave uncovered areas. It was observed that overlapping of 20 - 80 %, preferably 30 - 50 %, of the coating trails will result in a faster levelling and improved coating uniformity, however, higher overlapping will apply more coating solution to the printing cylinder and result in higher coating weights. The width of the coating trails is depending on the coating gap and the coating roll rotation. The smaller the gap and the faster the coating roll rotation, the broader the width of the coating trail. Therefore, all four settings will influence uniformity and coating weight of a specific coating solution.

[0037] High coating accuracy from start to end and high sleeve-to-sleeve reproducibility can be achieved by the new coating technology due to the possibility to use a sensor positioning system of fibre optic sensors which measure printing cylinder dimensions, printing cylinder diameter, printing cylinder position and coating roll position with high precision. With these data the machine software adjusts coating parameters, printing cylinder rotation and coating table speed for constant coating weight. Precision gap control for roller position is provided with good repeatability. In addition to the software controlled gap adjustment a mechanical gap adjustment is part of the coating head. This feature is used for gap calibration, gap re-adjustment or if the software gap setting needs a correction. It makes new gap settings or changing of gap settings convenient and easy. No new software programming is required if the gap distance needs to be changed for a single coating process or a special coating adjustment. The mechanical adjustment is performed by a micrometer which allows changes between coating roll and printing cylinder surface with an accuracy better than 5 µm which is sufficient to achieve the desired coating uniformity. The position is displayed and can be monitored by a 3-digit read-out. A wide variety of printing cylinders with different diameters and lengths can be used with the coating apparatus of the present invention..

Industrial Utility

[0038] The printing cylinder coated by the process of the present invention is directly ready for further processing. In case that photopolymerizable printing cylinders or sleeves have been coated with an infrared sensitive layer, such processing usually comprise the steps of imagewise exposure of the infrared sensitive layer, overall exposure with actinic radiation of the photopolymerizable layer through the imaged infrared sensitive layer, development with a suitable wash off solvent, drying, and post treatment. First, the infrared sensitive layer is exposed with an infrared laser, for example, a diode laser emitting between 750 and 880 nm, preferably 780 and 850 nm, or a YAG laser emitting at 1060 nm. The optional-strippable cover sheet may be removed prior to the laser exposure, in which case the laser 5 vaporizes the infrared sensitive layer. If the coversheet remains on the photopolymerizable printing cylinder, the exposure by laser removes the infrared sensitive layer to the overlying coversheet and is stripped off upon removal of the coversheet. The photopolymerizable printing cylinder is exposed overall with conventional radiation sources, such as, for example, xenon lamps, carbon arc lamps, mercury vapor lamps, fluorescent lamps having phosphors emitting UV 10 radiation, etc. The unpolymerized areas can be washed off, depending on the binder system, with water, aqueous or semi-aqueous solutions, or suitable organic developer solvents, such as, for example, aliphatic or aromatic hydrocarbons, terpenes, toluene, halogenated hydrocarbons, etc., or mixtures of the named solvents. Additives, such as surfactants or alcohols are possible. This step removes the unphotopolymerized areas of the photopolymerizable printing 15 cylinder, the remaining areas of the infrared sensitive layer, and a barrier layer that may optionally be present. After drying, the resulting flexographic printing cylinder can be post-exposed and/or chemically or physically treated in any sequence to prepare a non-tacky printing surface. These process steps are thoroughly described in WO 94/038383 or WO 94/03839. Continuous printing forms made by this process have applications in the flexographic printing of 20 continuous designs such as in wall-paper, decoration and gift wrapping paper.

EXAMPLES

[0039] The following examples illustrate the invention, but do not limit it. The average molecular weights of the polymers are given as weight average (Mw).

Example 1

[0040] A coating solution of an infrared sensitive material was prepared in the following way:

a solvent soluble thermoplastic polyamide resin with a softening point of about 140 °C and
an average molecular weight Mw of 20,000 was dissolved in a solvent blend with a high-shear dissolver. The
dissolver disk rotated with a tip speed of 16 m/sec. A carbon black pigment was added and dispersed into this
polymer resin solution. The concentration was adjusted to 36 % by weight of total solids. This polymer/pigment
dispersion was milled in a media mill in 4 passes at a mill base throughput of 90 kg/h. After milling the pigment
concentrate was diluted under high-shear with the dissolver. During diluting, coating additives were added in the
following sequence: a defoaming additive, a substrate wetting additive and a viscosity builder. The concentration
was adjusted to 4.8 % by weight of total solids.

[0041] This coating solution was filled into the container of the coating machine so that the coating roll dipped with its lower part to about 40% into the coating fluid. The machine was set for the following coating conditions: printing cylinder rotation 30 rpm, linear coating head speed 17.5 cm/min, coating roll rotation 21.5 rpm. A printing cylinder was used covered with a thin seamless layer of Cyrel® HORB (E.I du Pont de Nemours & Company, Wilmington, Del) photopolymerizable material.

[0042] A coating roll A was used made from solid Nylon®, diameter 110 mm, width 28 mm, roundness accuracy about 0.01 mm, surface roughness Rz = 1.6/1. The coating was applied in a way that the coating roll did not touch the surface of the printing cylinder (non-impact coating mode); the gap distance was adjusted to 60 µm between the tacky HORB surface and the Nylon® coating roll. The coating was dried with heated air of 26°C and relative humidity of 29 %. The coating was rub-resistant after about 40 min. A very uniform highly glossy black layer was achieved with no coating defects. The layer was peeled off with a clear tape and the optical density (OD) profil was measured with a transmission densitometer in coating direction parallel to the axis of the printing cylinder. Also the dry coating weight was determined by lifting and peeling off the photomask layer from the HORB photopolymerizable layer.

Results for coating solution coated with coating roll A and gap adjustment of 60 µm	
measured coating weight along the OD profil.	28 mg/ dm ²

(continued)

Results for coating solution coated with coating roll A and gap adjustment of 60 µm	
measured average OD along the OD profil	3.12
min / max OD	3.05 / 3.29
difference max - min OD	0.24
levelling of coating	complete levelling
coating spiral structure	none
mechanical scratches on the photopolymerizable layer	none

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[0043] The prepared infrared sensitive layer showed excellent coating quality and OD uniformity. The infrared sensitive layer was laser imaged with an Nd:YAG laser at 1064 nm. After processing a printing cylinder for high-quality flexographic printing was obtained.

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Comparative Example 1

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[0044] In a second experiment, the coating solution of Example 1 was used with the same coating and drying conditions as given in Example 1. The printing cylinder had the same geometries and was prepared in the same way as described in Example 1, also covered with Cyrel® HORB photopolymerizable material. A coating roll B was used made from flexible foamed polyurethane, diameter 110 mm, width 28 mm. This coating roll was adjusted in a way that the roll was pressed against the unexposed and tacky photopolymerizable surface; the flexible roll was deformed and the deformation was adjusted to about 1 mm displacement (impact coating mode).

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Results for coating solution coated with coating roll B and displacement of 1 mm	
measured coating weight along the OD profil.	31 mg/dm ²
measured average OD along the OD profil	3.28
min / max OD	2.64 / 3.69
difference max - min OD	1.05
levelling of coating	insufficient levelling
coating spiral structure	yes
mechanical scratches on the photopolymer	yes

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[0045] The infrared sensitive layer showed the spiral coating structure of the coating trails; a glossy trail alternated with a matte trail. The levelling was insufficient. The layer showed significant differences for the measured OD: highest 3.69, lowest 2.64. An unacceptable high difference of optical density ($\Delta = 1.05$) was measured. Due to mechanical scratches from the coating roll on the surface of the unexposed, tacky photopolymerizable layer, the printing cylinder could not be used.

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Claims

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1. A process for coating a printing cylinder with a layer of a liquid, comprising the steps of:

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- (a) forming a fluid film of the liquid on a surface of a coating roll;
- (b) positioning the surface of the coating roll in a predetermined distance from an outer surface of the printing cylinder such that the fluid film contacts the outer surface and a coating gap between the outer surface of the printing cylinder and the surface of the coating roll is formed;
- (c) simultaneously rotating and moving the coating roll relative to the printing cylinder in such a manner that the printing cylinder is coated with the liquid layer; and
- (d) drying the liquid layer to form the coated printing cylinder.

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2. The process according to claim 1,
characterized in that the printing cylinder is rotationally supported at both ends.

3. The process according to claims 1 - 2,

characterized in that the fluid film on the top of the coating roll is split into two parts according to adjust coating thickness.

4. The process according to claims 1 - 3,
5 characterized in that the fluid film split into two parts is adjusted by varying the coating gap between rotating printing cylinder and coating roll.
5. The process according to claims 1 - 4,
10 characterized in that the width of the predetermined distance between printing cylinder and coating roll is about 30 - 800 µm.
6. The process according to claims 1 - 5,
15 characterized in that the coating roll is rotated and moved from one end of the printing cylinder toward the other end of the printing cylinder in such a manner that a uniform overlapping spiral of the fluid film of the liquid is formed on the printing cylinder.
7. The process according to claim 6,
characterized in that the trails of the overlapping spiral of the fluid film have an overlapping of 20 - 80 %.
- 20 8. The process according to claims 1 - 7,
characterized in that the printing cylinder comprises a photopolymerizable elastomeric printing layer.
9. The process according to claims 1 - 8,
25 characterized in that the printing cylinder comprises a photopolymerizable elastomeric printing layer cylindrically formed on a sleeve.
10. The process according to claims 1 - 9,
characterized in that a hard coating roll is used.
- 30 11. The process according to claims 1 - 10,
characterized in that the material of the coating roll has a Shore D hardness measured according to ASTM D 2240 of at least 60.
12. The process according to claims 1 - 11,
35 characterized in that the coating roll material is selected from the group consisting of thermoplastic or thermo-setting non-elastomeric polymers, metals, and ceramic materials.
13. The process according to claims 1 - 12,
40 characterized in that the coating roll material is selected from the group consisting of polyamides and polyesters.
14. The process according to claims 1 - 13,
characterized in that the coating roll has a roll diameter of 90 - 150 mm.
- 45 15. The process according to claims 1 - 14,
characterized in that the coating has a roll width of 10 - 40 mm
16. The process according to claims 1 - 15,
characterized in that the liquid is an infrared-sensitive composition.
- 50 17. The process according to claims 1 - 16,
characterized in that the liquid is an infrared-ablative composition.
18. The process according to claim 17,
characterized in that the infrared-ablative composition comprises carbon black
- 55 19. The process according to claims 1 - 18,
characterized in that the process steps (a) to (d) are repeated at least once.

20. The process according to claim 19,
characterized in that for each repeat of the process steps (a) to (d) different liquids are used.
21. An apparatus to perform the process according to claims 1 - 20 comprising:
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(A) means to support and rotate the printing cylinder,
(B) means to support, rotate, and drive the coating roll in a predetermined distance to the printing cylinder,
(C) means to control the predetermined distance between the rotating printing cylinder and the rotating and moving coating roll,
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(D) a container to provide the liquid,
(E) means to control and adjust drying conditions.
22. The apparatus according to claim 21,
characterized in that the container to provide the liquid is installed on a vertically and horizontally moveable table.
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23. The apparatus according to claims 21 - 22,
characterized in that fibre optic sensors are used as control means.
24. The apparatus according to claims 21 - 23,
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characterized in that the predetermined distance between the rotating printing cylinder and the rotating and moving coating roll is adjusted by a software controlled motor.
25. The apparatus according to claims 21 - 24,
characterized in that the predetermined distance between the rotating printing cylinder and the rotating and moving coating roll is adjusted mechanically.
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26. The apparatus according to claims 21 - 25,
characterized in that the predetermined distance between the rotating printing cylinder and the rotating and moving coating roll is adjusted by a micrometer.
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27. A photopolymerizable flexographic printing sleeve comprising an outermost layer which was applied by the process according to claims 1 - 20.

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EUROPEAN SEARCH REPORT

Application Number
EP 00 11 0644

DOCUMENTS CONSIDERED TO BE RELEVANT			
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The present search report has been drawn up for all claims			
Place of search	Date of completion of the search	Examiner	
THE HAGUE	6 October 2000	Haenisch, U	
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**ANNEX TO THE EUROPEAN SEARCH REPORT
ON EUROPEAN PATENT APPLICATION NO.**

EP 00 11 0644

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 The members are as contained in the European Patent Office EDP file on
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